# Los Alamos National Laboratory

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# **Passive Neutron Multiplicity Counter**

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## Safeguards Assay Group NIS-5

#### **MS E540**

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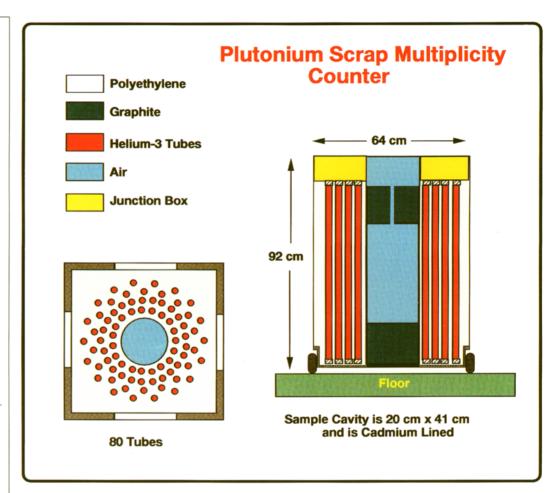


Fig. 1. In this cross section of the in-plant passive neutron multiplicity counter 126 helium-3 tubes (red) are arranged around the sample cavity. The space between the tubes is filled with polyethylene (white). The graphite above and below the sample cavity (green) scatters neutrons trying to exit the top and bottom of the cavity. The junction box (yellow) contains the amplifiers. The sample cavity is open to the air (blue) at atmospheric pressure.

assive neutron multiplicity counting is a nondestructive measurement technique developed to assay materials containing plutonium. Figure 1 illustrates the internal structure of a passive neutron multiplicity counter designed for pyrochemical studies. The technique is an extension of passive neutron coincidence counting that measures two quantities: the total neutron counting rate and the coincidence counting rate. Many plutonium samples have three major unknown quantities that affect the two counting rates: the plutonium mass, the neutron multiplication, and the neutron yield from (alpha, n) reactions. Because conventional coincidence counting only measures two quantities, either the neutron multiplication or the (alpha, n) yield must be known to determine the plutonium mass. For many impure or heterogeneous samples, neither the multiplication nor the (alpha, n) yield can be known beforehand, so the measurement of a third quantity is required.

Neutron multiplicity counting is one way to get a third quantity. The average number of neutrons emitted by a spontaneous fission in plutonium is approximately two, and the average number emitted by an induced fission in plutonium is approximately three. Multiplicity counting can be used to determine the neutron multiplication because the number of neutrons emitted in a burst following a spontaneous fission increases with increasing multiplication. The three measured quantities used for multiplicity counting are called the singles, doubles, and triples counting rates. The singles and doubles rates are the same as the totals and coincidence rates in conventional coincidence counting. The triples rate is new and measures a higher level of correlation in the neutron pulse stream. For analysis, the singles, doubles, and triples rates are expressed as functions of the <sup>240</sup>Pu spontaneous fission rate, the neutron multiplication, and the (alpha, n) yield. These three equations are then solved for the three unknowns; finally, the plutonium mass is determined from the <sup>240</sup>Pu mass and the isotopic composition as in conventional coincidence counting.

#### **Detectors and Electronics**

Neutron multiplicity counters are similar to conventional coincidence counters; they are based on the detection of neutrons with <sup>3</sup>He proportional counters embedded in a polyethylene moderator, and they use the same amplifier electronics system. There are three important differences however. First, these counters must have higher detection efficiencies than conventional counters because the triples rate is proportional to the cube of the efficiency. Typically, multiplicity counters have detection efficiencies ranging from 40 to 55%. To obtain such high efficiencies, 80 to 130 <sup>3</sup>He tubes are needed. Figure 1 shows the tube configuration for the in-plant (or pyrochemical) multiplicity counter. This configuration results in a 56% detection efficiency for the neutrons from the plutoniuim. Second, the electronics deadtimes for multiplicity counters must be small because the corrections for the triples rate can be large. To achieve low deadtimes, 20 to 55 amplifiers have been used. Third, the detection efficiency of a multiplicity counter must be less dependent on neutron energy. Figure 2 shows efficiency vs. energy for the in-plant multiplicity counter compared to a conventional high-level neutron coincidence counter (HLNC), which was designed for portable coincidence counting applications.

Special multiplicity electronics are required to measure the neutron multiplicity distributions in the coincidence gates. Multiplicity electronics provide the same data as a conventional shift-register coincidence module and, in addition, provide the multiplicity distributions. The multiplicity measurement records the number of times each multiplicity occurs in the coincidence gates. For example, if seven neutron pulses are in a coincidence gate when another neutron arrives, then "1" is added to the counter that tallies multiplicities of seven. A multiplicity distribution is measured for the real-plus-accidental coincidence gate and for the accidental coincidence gate. The accidental gate measures random coincidences that must be removed from the real-plus-

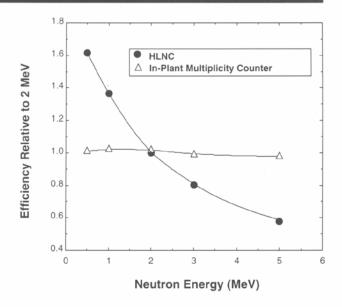


Fig. 2. Neutron detection efficiency vs. neutron energy for the in-plant multiplicity counter and the portable, high-level neutron coincidence counter (HLNC).

accidental coincidences to obtain the real coincidences. Table I shows the real-plus-accidental and the accidental multiplicity distributions for a 1-kg plutonium sample measured for 1000 s. As an example from this table, 20 neutron pulses were found in the real-plus-accidental coincidence gate 78 times.

## **Data Analysis**

The singles, doubles, and triples counting rates are calculated from the real-plus-accidental and accidental multiplicity distributions. Electronic deadtime corrections are required. The singles, doubles, and triples rates are related to the effective <sup>240</sup>Pu mass, the neutron multiplication, and the (alpha, n) neutron rate through three theoretical equations. The model assumes that the neutron detection efficiency is constant and the neutron multiplication is constant for all neutrons from the sample. To the extent that the theoretical model matches the plutonium samples, the measured singles, doubles, and triples rates determine the effective <sup>240</sup>Pu mass, the multiplication, and the (alpha, n) rate *without a calibration curve*.

Table I. M	ultiplicity Dis	stributions fo	r a 1000-s M	easurement (	of 1 kg of
T Iditollidin	Counts			Counts	
Multiplicity	Real + Accidental	Accidental	Multiplicity (cont.)	Real + Accidental (cont.)	Accidental (cont.)
0	4863880	5708122	13	103926	75358
1	16014347	17977225	14	40246	28536
2	27615893	29696698	15	15106	10416
3	33191194	34213111	16	5522	3736
4	31244355	30927348	17	1872	1193
5	24535179	23349181	18	702	440
6	16706604	15317903	19	216	136
7	10161183	8979892	20	78	52
8	5604155	4785248	21	21	9
9	2853120	2359023	22	10	- 5
10	1356495	1082439	23	6	1
11	606426	469055	24	2	0
12	257046	192488	25	0	0

There are two cases of practical importance where the theoretical model differs significantly from the measured samples, so that corrections are required. (1) If samples have high neutron multiplication, then the multiplication is not constant throughout the sample. However, when the dimensions of a sample are known approximately, a correction factor can be calculated to account for the variation of the neutron multiplication throughout the sample. (2) If samples are highly moderating, or if the energy of the (alpha, n) neutrons is much different from fission neutron energies, then the efficiency is not constant. Problems related to varying neutron energies are first minimized by designing the detector to have an efficiency that is insensitive to neutron energy. In addition, the ratio of counting rates in the inner and outer rings of <sup>3</sup>He tubes is sensitive to neutron energy and can be used to correct for variations in (alpha, n) neutron energies. The multiplicity electronics module contains two extra pulse counters to facilitate the measurement of ring ratios.

## **Applications**

Three multiplicity counters have been built to date: a research counter at Los Alamos, an in-plant counter for temporary use in DOE facilities, and a counter for the International Atomic Energy Agency (IAEA). See Fig. 3. The research counter is used primarily to develop multiplicity counting techniques. The in-plant counter has been used in the Los Alamos plutonium facility and at the Lawrence Livermore National Laboratory to assay inventory items. The IAEA counter is used for routine verification of mixed oxide scrap inventory items in Japan. A fourth counter (an active/passive multiplicity counter) will be permanently installed in the Los Alamos plutonium facility. Additional counters are being designed for domestic and international arms control and arms reduction applications.

Multiplicity counting provides a higher level of verification than is possible with conventional coincidence counting. For example, the multiplicity counter gives a direct measure of the neutron multiplication of a sample. The plot in Fig. 4 shows the triples/doubles ratio vs. the neutron multiplication; this curve is nearly independent of sample type. As an example, a 2-kg plutonium metal sample that has a multiplication of 2.0 can be distinguished easily from isotopic neutron sources with a multiplicity counter; this distinction is not possible with a conventional coincidence counter.

Conventional coincidence counters can be used as multiplicity counters with the addition of multiplicity electronics and software. The performance of such systems cannot match that of counters designed specifically as multiplicity counters but can be adequate in some circumstances—shipper/receiver confirmations, for example.

#### **Performance**

Figure 5 presents assay results obtained with the in-plant multiplicity counter. The data were analyzed with conventional coincidence counting techniques (using only singles and doubles rates) and with multiplicity counting techniques (using singles, doubles, and triples rates). The conventional tech-

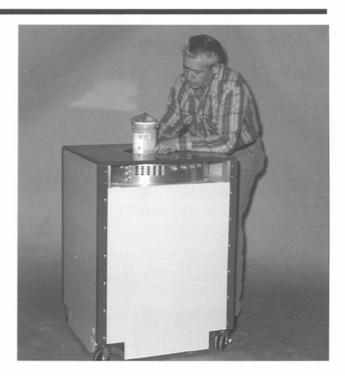


Fig. 3. Photograph of the plutonium scrap multiplicity counter used by the International Atomic Energy Agency for mixed oxide scrap verification measurements in Japan.

nique requires assumptions about the neutron multiplication or the (alpha, n) yield of the samples, so the samples were assumed to be pure. The impurities in the samples bias the conventional results high because fissions are induced by the excess (alpha, n) reactions, whereas the multiplicity analysis determines the (alpha, n) rate and corrects for it automatically.

The statistical precision of multiplicity measurements is strongly dependent on the sample type and mass. For 1-kg samples of plutonium metal and oxides with slight impurities, the precision in a 1000-s measurement is typically 0.3 to 2%. The multiplicity technique is not well suited to samples that

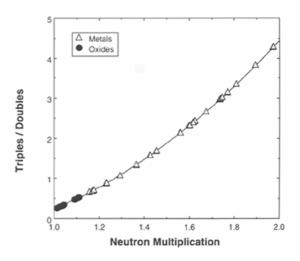


Fig. 4. Triples/doubles ratio vs. neutron multiplication.

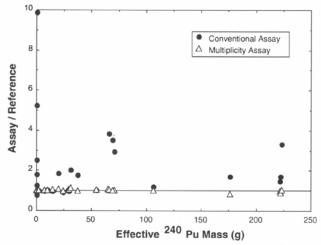


Fig. 5. Assay results from the in-plant counter analyzed with multiplicity and conventional techniques.

produce many more (alpha, n) neutrons than spontaneous fission neutrons because the errors from counting statistics are large for these samples.

Based on experience to date, the expected accuracy of multiplicity assay for cans of plutonium is ~2% for oxides and metals with low neutron multiplication and ~5% for metals with high neutron multiplication. More experience is required to determine the accuracy of multiplicity counting for a wide variety of sample types.

## **Procuring a Multiplicity Counter**

The multiplicity electronics module is now commercially available as a Nuclear Instrumentation Module (Model 2150) from Canberra Industries, Meriden, Conn. Software to perform multiplicity assays with the Model 2150 is available from the Safeguards Assay Group at Los Alamos. There are two choices for a multiplicity detector: add multiplicity electronics to an existing detector or build a new detector. The Los Alamos Safeguards Assay Group can advise on the suitability of existing counters for specific multiplicity applications and can assist with the procurement of custom multiplicity counters. This assistance can range from advice to design, fabrication, software support, calibration, evaluation, documentation, and training.

### **Additional Sources of Information**

- M. S. Krick and J. E. Swansen, "Neutron Multiplicity and Multiplication Measurements," Nucl. Instr. Meth. 219, 384-393 (1984).
- K. Boehnel, "The Effect of Multiplication on the Quantitative Determination of Spontaneously Fissioning Isotopes by Neutron Correlation Analysis," *Nucl. Sci. Eng.* 90, 75–82 (1985).
- D. M. Cifarelli and W. Hage, "Models for a Three Parameter Analysis of Neutron Signal Correlation Measurements for Fissile Material Assay," *Nucl. Instr. Meth.* A251, 550 (1985).
- N. Ensslin, N. Dytlewski, and M. S. Krick, "Assay Variance as a Figure of Merit for Neutron Multiplicity Counters," *Nucl. Instr. Meth.* A290, 197–207 (1990).
- N. Dytlewski, "Dead-time Corrections for Multiplicity Counters," Nucl. Instr. Meth. A305, 492–494 (1991).
- D. G. Langner, et al., "Neutron Multiplicity Counter Development," 13th ESARDA Symp. (ESARDA, Liege, Belgium, 1991).
- J. K. Halbig, et al., "Recent Developments in Multiplicity Counting Hardware at Los Alamos," IEEE Nucl. Sci. Symp., Santa Fe, New Mexico, November 1991.
- N. Dytlewski, M. S. Krick, and N. Ensslin, "Measurement Variances in Thermal Neutron Coincidence Counting," *Nucl. Instr. Meth.* A327, 469–479 (1993).
- H. O. Menlove, et al., "Plutonium Scrap Multiplicity Counter Operation Manual," Los Alamos National Laboratory report LA-12479-M (January 1993).
- M. S. Krick and W. C. Harker, "Multiplicity Neutron Coincidence Counting User's Manual," Los Alamos National Laboratory document LA-UR-93-1394 (1993).
- D. G. Langner, et al., "Thermal Neutron Multiplicity Measurements Using the Pyrochemical Multiplicity Counter at Lawrence Livermore National Laboratory," INMM Meeting, Scottsdale, Ariz., July 18–21, 1993 (LA-UR-93-2610).

